

Photon coincidences in spontaneous parametric down-converted radiation excited by a blue LED in bulk LiIO_3 crystal

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Abstract: We report on experimental and numerical investigation of two-photon coincidence properties of the parametric spontaneous down-converted field excited by a high brightness blue LED in bulk lithium iodate crystal. Ratio of up to 11.5% of coincidence, which cannot be attributed to classical coincidences, to single photon counts was recorded at the outputs of multimode fibers, demonstrating well-preserved biphoton property. This result, combined with practically useful power of the source, suggests its possible application for a class of quantum experiments.

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References and links

1. W. H. Louisell, A. Yariv, and A. E. Siegman, "Quantum fluctuations and noise in parametric processes," *Phys. Rev.* **124**(6), 1646–1654 (1961).
2. S. E. Harris, M. K. Oshman, and R. L. Byer, "Observation of tunable parametric fluorescence," *Phys. Rev. Lett.* **18**(18), 732–734 (1967).
3. A. A. Malygin, A. N. Perin, and A. V. Sergienko, "Efficient generator of a two-photon field of visible radiation," *Sov. J. Quantum Electron.* **11**(7), 939–941 (1981).
4. C. K. Hong, Z. Y. Ou, and L. Mandel, "Measurement of subpicosecond time intervals between two photons by interference," *Phys. Rev. Lett.* **59**(18), 2044–2046 (1987).
5. J. W. Pan, Z.-B. Chen, M. Żukowski, H. Weinfurter, and A. Zeilinger, "Multi-photon entanglement and interferometry," *arXiv:0805.2853v1*, <http://arxiv.org/abs/0805.2853>.
6. M. D'Angelo, M. V. Chekhova, and Y. Shih, "Two-photon diffraction and quantum lithography," *Phys. Rev. Lett.* **87**(1), 013602 (2001).
7. T. Scheidl, R. Ursin, A. Fedrizzi, S. Ramelow, X.-S. Ma, T. Herbst, R. Prevedel, L. Ratschbacher, J. Kofler, T. Jennewein, and A. Zeilinger, "Feasibility of 300 km quantum key distribution," *N. J. Phys.* **11**(8), 085002 (2009).
8. M. A. Horne, A. Shimony, and A. Zeilinger, "Two-particle interferometry," *Phys. Rev. Lett.* **62**(19), 2209–2212 (1989).
9. J. Brendel, E. Mohler, and W. Martienssen, "Experimental Test of Bell's Inequality for Energy and Time," *Europhys. Lett.* **20**(7), 575–580 (1992).
10. P. G. Kwiat, K. Mattle, H. Weinfurter, A. Zeilinger, A. V. Sergienko, and Y. Shih, "New high-intensity source of polarization-entangled photon pairs," *Phys. Rev. Lett.* **75**(24), 4337–4341 (1995).
11. A. Mair, A. Vaziri, G. Weihs, and A. Zeilinger, "Entanglement of the orbital angular momentum states of photons," *Nature* **412**(6844), 313–316 (2001).
12. J. T. Barreiro, N. K. Langford, N. A. Peters, and P. G. Kwiat, "Generation of hyperentangled photon pairs," *Phys. Rev. Lett.* **95**(26), 260501 (2005).
13. D. L. Weinberg, "Observation of Optical Parametric Noise Pumped by a Mercury Lamp," *J. Appl. Phys.* **41**(10), 4239–4240 (1970).
14. A. Politi, J. C. F. Matthews, and J. L. O'Brien, "Shor's quantum factoring algorithm on a photonic chip," *Science* **325**(5945), 1221 (2009).
15. G. Tamošauskas, J. Galinis, A. Dubietis, and A. Piskarskas, "Observation of spontaneous parametric down-conversion excited by high brightness blue LED," *Opt. Express* **18**(5), 4310–4315 (2010).
16. P. G. Kwiat, K. Mattle, H. Weinfurter, A. Zeilinger, A. V. Sergienko, and Y. Shih, "New high-intensity source of polarization-entangled photon pairs," *Phys. Rev. Lett.* **75**(24), 4337–4341 (1995).

17. X. Y. Zou, L. J. Wang, and L. Mandel, "Violation of classical probability in parametric down-conversion," *Opt. Commun.* **84**(5-6), 351–354 (1991).
 18. P. S. K. Lee, M. P. van Exter, and J. P. Woerdman, "How focused pumping affects type-II spontaneous parametric down-conversion," *Phys. Rev. A* **72**(3), 033803 (2005).
 19. S. Cialdi, F. Castelli, and M. G. A. Paris, "Properties of entangled photon pairs generated by a CW laser with small coherence time: theory and experiment," *J. Mod. Opt.* **56**(2), 215–225 (2009).
 20. J. Galinis, G. Tamošauskas, and A. Piskarskas, "Modeling of photon coincidence and dispersive properties of spontaneous parametric down-converted field excited by incoherent source," in preparation.
 21. J. G. Rarity, K. D. Ridley, and P. R. Tapster, "Absolute measurement of detector quantum efficiency using parametric downconversion," *Appl. Opt.* **26**(21), 4616–4619 (1987).
 22. Z. Zhao, K. A. Meyer, W. B. Whitten, and R. W. Shaw, "Optical absorption measurements with parametric down-converted photons," *Anal. Chem.* **80**(19), 7635–7638 (2008).
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1. Introduction

The discovery of spontaneous parametric down-conversion (SPDC) [1,2] came in fruition of a variety of studies. SPDC has been recognized as an attractive two-photon source [3], later used in the famous Hong-Ou-Mandel two-photon interference experiment [4], and since then employed in many fundamental [5] and applicative studies [6,7]. It has the advantage of being an inexpensive, relatively simple to use tool, able to produce pairs of photons correlated in path [8], time [9] and other degrees of freedom [10–12].

Quite early, some considered stimulating SPDC by an incoherent pump source. The very first experimental demonstration [13] of an SPDC pumped by mercury lamp atomic line emission was designed to imitate radiation of the laser by both time and spatial coherence. Due to their large accessibility, low price, and reasonably high output power and broad range of wavelengths available, incoherent light sources, used as pump for SPDC, can play an important role in construction of complex photonic quantum chips designed e.g. for quantum computation [14]. Recently, SPDC has been demonstrated to occur in nonlinear crystals when pumped by incoherent light of a high-brightness broadband blue LED [15]. LEDs almost uniformly cover wavelength range from 260 nm to 4600 nm as provided by Thorlabs, Inc., for example. It was shown, both experimentally and analytically, that the coherence of the pump is not an obligatory factor for SPDC to take place, thanks to the linear behavior of the process when excitation can be decomposed into independent power portions and the result expressed as the sum of SPDC powers generated individually. The few existing experimental demonstrations [13,15] did not analyze the quantum properties of the down converted field pumped by an incoherent source but only demonstrated the detectable power of SPDC. Incoherent pump results in rather chaotic spatial distribution of SPDC lowering coincidence probability at spatially narrowband detection. Moreover, possibility for pairs to pass frequency-narrowband filter typically used in quantum experiments diminishes due to the extended frequency bandwidth of the LED pump. Poor spatial coherence of the pump leads to very low radiance of excitation. Comparing the parameters of the LED to the laser used in high intensity entangled photon source (e.g [16].), the latter is of approximately 10 orders of magnitude higher spectral radiance. It is known that spectral radiance of SPDC is linked to spectral radiance of the pump at optimized conditions. Additionally, detection capabilities of convenient photon counters are proportional to the spectral radiance of the source when interference filters are used. As a result, biphoton property is not necessarily preserved and detectable. Experiment might answer, whether such source is suitable for quantum experiments where biphoton property is the key factor. The question is related not to the origin of radiation but to capability of the incoherent pump to deliver detectable flux of biphotons among accidental coincidences that inevitably take place at high flux of singles [17].

Quantum properties of SPDC as of non-classical field have been well proven in numerous experiments, typically employing the Hong-Ou-Mandel effect [4]. Investigation and confirmation of non-classical behavior of the observed radiation is out of scope of the present research. To the best of our knowledge, SPDC is the only process capable of sourcing biphotons in employed experimental realization. The goal of this experiment was to perform characterization of the light source itself – to investigate dependencies of biphoton and single

photon fluxes on parameters of the exciting field. As a result, we omit discussion on the origins of radiation and the comparison with other biphoton sources and possible manipulations exploiting non-classical behavior.

Previous works were attributed to the investigation of the influence of separately spatial [18] and frequency [19] bandwidth of the pump, whereas laser pumped experiments supported the analysis. Theoretical analyses based on semiclassical model [15,20] are related only to radiant power and spectral radiance calculations. We adopted the semiclassical model for calculation of coincidence property [20] and obtained satisfactory match with the results of the experiment.

The first experimental investigation of quantum properties of SPDC excited by the continuous wave LED demonstrates up to 11.5% photon count coincidences when possible coincidences according to the classical field statistics are excluded. The present value attributes to biphoton content, thus indicating non-classical origin of the excited radiation. This opens perspectives for employing incoherently pumped SPDC sources in a class of quantum experiments where a field of high spatial coherence is not a crucial prerequisite, as in several metrological applications [21,22]. The obtained results are analyzed numerically using an extended semiclassical model [20], thus verifying its capability of calculation of photon coincidence properties in broadband biphoton field excited by a both spatially and frequency broadband pump.

2. Experimental setup and numerical model

The experimental setup (Fig. 1a) was designed utilizing the traditional photon coincidence counting arrangement with two photon-counting detectors coupled to the photon source by fibers. Pump source design is based on a high brightness mass production light emitting diode LZ1-10UA05 from LedEngin, Inc. (LED in Fig. 1a). Total emission power of the LED is 0.9 W, peak wavelength 403.5 nm and bandwidth 14.4 nm FWHM. The rest of the pump source setup is dedicated to beam shaping and imaging: lenses $L1$ and $L2$ form the beam waist; aperture $A1$ controls spatial bandwidth; $A2$ controls the beam diameter; lenses $L3$ and $L4$ image the waist of the beam formed at the aperture $A2$ into the center of nonlinear crystal $LiIO_3$; Glan prism $GLAN$ supplies polarization control, which is also useful for background counts measurements. The pump beam maintains almost uniform flattop shape and diameter while propagating inside the nonlinear crystal due to short wavelength, even if its divergence is far from limited by diffraction.

Lithium iodate crystal of 20 mm length, cut at $\theta = 35^\circ$ for Type I interaction, is used for generation of the down-converted radiation. The crystal is oriented at $\theta = 43.4^\circ$ with respect to the pump. SPDC radiation around degeneracy wavelength emerges as a 0.1 rad opening angle cone. Two identical channels for registration of SPDC radiation are mounted transversely in a non-critical ϕ plane at ± 50 mrad angles. They consist of interference filters $IF1$, $IF2$ with 37.5 nm bandwidth around degeneracy (Thorlabs, Inc. FB800-40), antireflection coated aspheric lenses $L5$, $L6$ with focal length $f = 11$ mm, multimode step refraction index fibers $FB1$, $FB2$ with core diameter of 105 μm and numerical aperture 0.22 (both from Thorlabs, Inc.) and a pair of PerkinElmer Inc. photon counters SPCM-AQRH-14-FC. TTL pulses from both photon counters are directed to a coincidence circuit (with coincidence window of 7 ns) and subsequently counted. Excitation frequency FWHM bandwidth is 1.3 times narrower than the detection bandwidth whose center has 1% offset with respect to degeneracy.

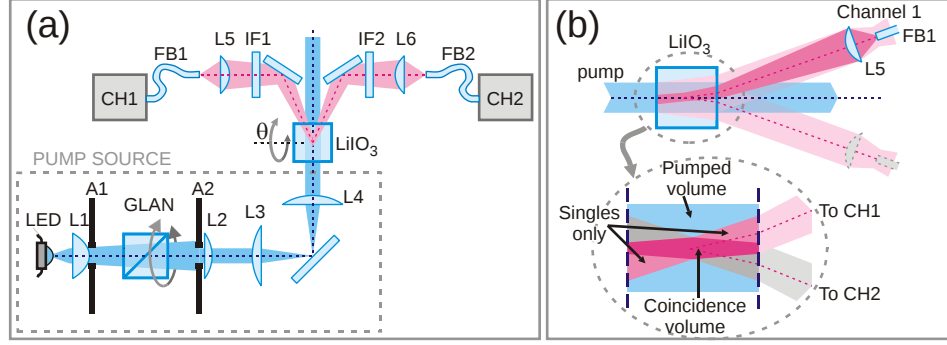


Fig. 1. A simplified scheme of experimental photon counting setup (a). Implemented geometrical model for computation of coupling SPDC radiation into fibers (b).

Numerical analysis is based on the semiclassical model described in details in [15,20]. Its capabilities were extended by implementing the coincidence probability calculation. The extended model is only suitable to compute power ratios and coincidence probabilities attributed to a particular spatial and wavelength bandwidth of both pump and SPDC. Due to these limitations, several assumptions need to be made in order to reproduce the experimental conditions of the present research. Firstly, the use of multimode fibers allowed us to simplify the wave propagation model down to a ray propagation model, similarly to the commonly used ray-tracing strategy. Secondly, properties of SPDC (mainly spatial bandwidth) fed into the fiber vary slightly along the crystal because the distance changes with respect to the lenses $L5$ and $L6$. However, this variation is relatively small compared to the acceptance spatial bandwidth itself. Therefore, we assume that SPDC properties are nearly identical along the crystal length. This assumption allows us to treat emission properties from the pumped channel along the crystal as uniform with respect to detection. Consequently, it allows us to separate the final SPDC flux expression into two independent components: an SPDC elementary volume emission multiplied by the geometry factor of the experiment.

Calculations are made for given power P , frequency ω_p and spatial spectrum (ϕ, θ) of the pump using second order nonlinear coefficient $d_{31} = 7.2$ pm/V. SPDC photon flux for crystal length L and given detection solid angle ρ_{det} is calculated from the following expression:

$$N_s = \int_0^\infty \int_0^{\rho_{\text{det}}} \int_0^{2\pi} \frac{\beta L^2}{\hbar \omega_s} P(\omega_p, \phi, \theta) q(\omega_s) \text{sinc}^2\left(\frac{\Delta k L}{2}\right) d\phi d\rho d\omega_s;$$

where β – nonlinear conversion coefficient, $q(\omega_s)$ – detection quantum efficiency, Δk – wave vector mismatch between pump, signal and idler fields, ϕ, ρ – detection angular coordinates. Solid angle is determined by the numerical aperture of the fiber divided by the magnification of the lens ($L5$ in Fig. 1b). Photon flux at the channel N_j ($j = 1, 2$ is channel number) is calculated by multiplying SPDC photon flux N_s by geometry factor for a channel (top image in Fig. 1b) which is equal to intersection of the pumped crystal volume and the volume fed into the fiber divided by the pumped crystal volume:

$$N_j = \frac{V_j \cap V_{\text{pump}}}{V_{\text{pump}}} \cdot N_s; \cap \text{attributes to intersection.}$$

Coincidence rate is calculated as follows. Assume that the first detector is used for gating. Flux of conjugated photons at the second detector $N_2(N_1)$ is obtained as the idler whose frequency is defined by the energy conservation law ($\omega_i = \omega_p - \omega_s$), taking into account bounds set by spatial and frequency acceptance bandwidth of the second channel. Finally, coincidence count rates are expressed as a paired photons flux $N_2(N_1)$ multiplied by the coincidence geometry factor – intersection of volumes fed into the fibers divided by the volume of coincidence count triggering channel $CC = \frac{V_1 \cap V_2}{V_1} \cdot N_2(N_1)$.

Spatial walk-off of the extraordinary polarized pump, which is important at small pump diameters, is taken into account in order to complete the geometrical model. Presented strategy was found to be of moderate accuracy for both qualitative and quantitative relations, in comparison with the experiment. There are several known limitations of the model that lead to its limited accuracy. First of all, spatial acceptance of the fiber is assumed to be perfectly flat-top-shaped, while in reality it is not. The same is valid for the transmittance function of the interference filter. These factors could be implemented into the existing model but this would result in drastically increased computation time because calculation for each combination of parameters should be made individually. Ray propagation model is clearly limited for the simulation of single mode fiber coupling. This is also linked to the limited accuracy of the simulation of a long crystal when high numerical aperture optics is used. Only the plane at the focus located in the center of the crystal is well imaged into the fiber while out-of-focus front and back planes are blurred. This effect is not taken into account in the model. The developed model, due to its simplified structure, is a compromise on accuracy, yet its major advantages are simple implementation and high computation speed.

The capability of the present model in precise power distribution calculations was tested in [15], therefore it was assumed to be accurate enough for calculating the SPDC properties of elementary volume. Overall accuracy was tested by comparing experimental and simulated photon count results for perpendicular scanning of the *FB1* fiber tip (Fig. 2a) and their dependence on crystal angle θ (Fig. 2b).

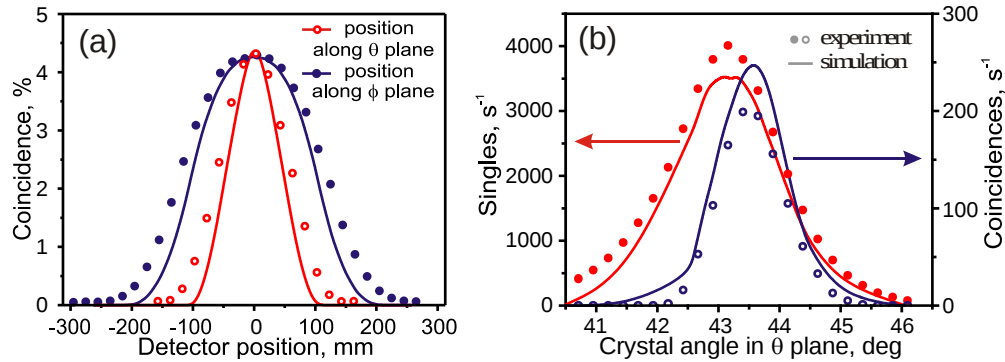


Fig. 2. Photon coincidence ratio dependence on fiber tip position in channel 1: experimental data (circles) and normalized numerical traces (a). SPDC photon flux at a single channel and coincidence rate dependence on crystal angular tuning (b).

Spatial overlap of the volumes imaged into fibers, which can be obtained without calculating SPDC output at all, is the only important parameter for the simulation of the fiber tip scanning, because SPDC properties of the elementary volume are assumed to be the same and the pump is uniform in the integrated volume. Experimental points and normalized simulation curves of the coincidence to single counts' ratio for the pump of 5 mm diameter and 25 mrad spatial spectrum show good qualitative and moderate quantitative (with numerically obtained 7% coincidence ratio) agreement.

Simulation of rotation of the nonlinear crystal in the θ plane, contrary to the fiber tip scanning, is only a function of the elementary volume properties, because the geometry of the detection channels is fixed. Results obtained for 5 mm diameter and 9 mrad spatial spectrum of the pump (Fig. 2b) show good both qualitative and quantitative agreement of the simulation and the experiment.

We can conclude that simple geometrical relations illustrated in Fig. 1b well describe SPDC properties in the presented experimental configuration. Overall SPDC properties, thanks to its linear behavior with respect to the pump power, can be obtained by combining semiclassical SPDC plane wave model [15,20] with a separately obtained spatio-geometrical model.

3. Coincidence measurements

The main target of the present work was experimental characterization of the properties of SPDC excited by incoherent source by means of photon coincidence measurements. The ratio of biphoton states is a widely used parameter for SPDC characterization. Therefore, we chose the standard two detector arrangement for photon coincidence measurement at variable parameters of the pump beam and fixed parameters of the detection channels. Low radiance of the process caused by the multimode pump had been expected in the experiment preparation stage. Therefore, it was only possible to use multimode fibers for spatial filtering and signal guiding to detectors.

The pump beam parameters: diameter and spatial bandwidth, were changed gradually: 0.8, 1.8, 3, 5, 8 mm and 9, 15, 25, 40 mrad respectively using two sets of hard apertures. The LED operated at constant power, but crystal pump power varied from 1.64 μW to 3.26 mW because the majority of power was lost in the beam shaper according to the constant spectral radiance law. The range of the beam sizes used was bound by the level of a well detectable signal, uniformity of the beam shape along the crystal and by the size of the area imaged into fibers. The lower limit of spatial bandwidth was also defined by the detection efficiency, while the upper limit by the acceptance angle of the Glan polarizer.

The main results of the experiment are the measured dependences of the photon flux at a channel (single counts) and coincidence count rate on the diameter and spatial spectrum of the pump beam. Single counts rates are shown with subtraction of the dark counts and background fluorescence measured with the same σ polarized pump power. Coincidence count rates are shown with subtraction of accidental coincidences that are equal to expected coincidences, in case all singles attribute to classical field. Their comparison to numerically simulated results is shown in Fig. 3. Numerical traces show progress of a single parameter such as beam diameter (green and red) or spatial bandwidth (blue and cyan) linked to the power of the constant radiance pump. Experimental points correspond to the same parameters as of the numerical simulation. For example, the bottom squares in Fig. 3a and b are the results of the measurement of 9 mrad spatial bandwidth pump with the expansion of the beam diameter from 0.8 mm to 8 mm.

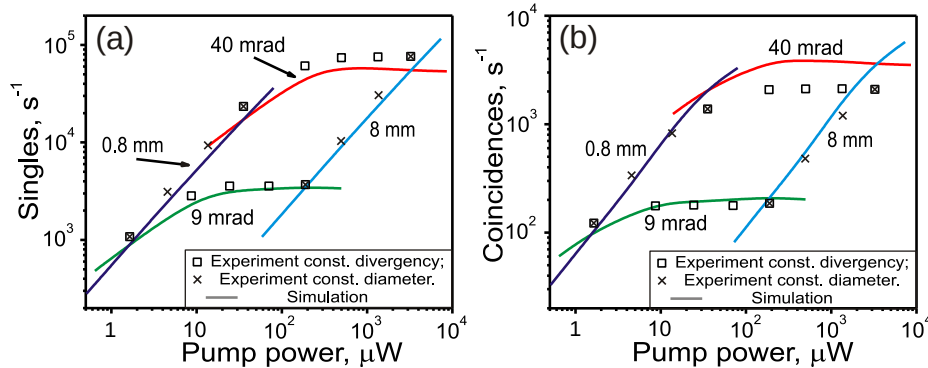


Fig. 3. Photon flux (a) and coincidence count (b) dependence on the pump beam diameter and divergence. Experimental points represent series of measurements performed with a pump of variable spatial spectrum 9, 15, 25, 40 mrad with constant beam size (×) and variable beam diameter 0.8, 1.8, 3, 5, 8 mm with constant spatial spectrum (□). Numerical simulation traces (solid lines) are obtained for respective fixed parameter of the pump of constant radiance.

The confirmation that SPDC power is sufficient for single photon and coincidence counting in multimode detection setup is a promising result for applications. For all the pump beam parameters both the coincidence and single counts significantly exceeded the respective accidental coincidence and dark count rates. Coincidence rate was 45 times higher than accidental coincidences for the worst case of single count rates exceeding $5 \cdot 10^4 \text{ s}^{-1}$, whereas single counts exceeded dark counts elevenfold in the case of the weakest 1.64 μW pump.

Classical field statistics violation ratio was estimated in order to check whether the detected coincidence counts come from a non-classical process. Estimation of the highest classical coincidence rates was made assuming that the classical source emits from the crystal. Parts of radiation recorded by two spatially separated detectors must follow the Poissonian photon number distribution because of the large frequency bandwidth of the detected radiation and its high number of spatial modes, supported by the fact that we are using a continuous wave pump. Classical probability violation ratio [17] $CC/CC_{\text{classical}}$ was obtained by dividing the experimentally recorded coincidence number CC by the highest expected coincidence number of the classical field $CC_{\text{classical}}$, as if it was recorded during the same amount of time as in the experiment. Coincidence window time, dead time of the photon counter, dark counts and errors of the values were taken into account for this evaluation calculating worst-case lowest values. Coincidence statistics do not contradict classical predictions when the presented ratio does not exceed 1. Obtained violation ratios ranged from 44 for the most spatially broadband and largest beam pump, to $9.8 \cdot 10^3$ for the smallest diameter and narrowest spatial spectrum of the pump, indicating non-classical origin of the detected radiation.

Single counts show excellent match to the second order dependence on the spatial bandwidth of the pump, whose power increases according to the same law. Coincidences have signs of saturation for the broadest spatial bandwidth of the pump, which matches with the simulation results as well (blue and cyan traces in Fig. 3b). The pump is still very effective even at such broad divergence. Both singles and coincidences show similar dependence on the pump beam diameter. Rates increase with increased beam diameter at the beginning and do not change anymore when the pump beam size is several times larger compared to the 1.05 mm diameter area imaged into fibers.

The best recorded 11.5% coincidences to singles ratio was observed at the smallest beam diameter and divergence. The geometry model of the experiment can easily explain the result, because the smallest pump beam of 0.8 mm diameter best fits into the region imaged into both fibers. Regions where only singles can be detected (see Fig. 1b) remain unpumped. Ratio of biphoton states in the SPDC radiation at the exit of the crystal was estimated to be 49% in the region which can be guided into multimode fibers (if regression due to losses in filters and fiber couplers as well as quantum efficiency of photon counters would be excluded). The present ratio would be even larger if the interference filters were perfectly centered at degeneracy.

Coincidence ratio drops from 5% at the smallest divergence almost linearly down to 2.8% at the highest divergence when the largest 5 mm and 8 mm pump beams produce uniform illumination. Qualitative description of the observed diminishing is complicated because of several factors taking place simultaneously. They include emission cone direction and cone opening angle variation with respect to different spatial components. Additionally, the coincidence ratio decreases when pump components phase-matched at fibers' direction are capable of producing only single photons in detection band, whereas paired ones are out of the detection band.

Summarizing the experimental part, it was demonstrated that an incoherent source, in this case a LED, is capable of exciting a biphoton field of moderate quality, which is well detectable in a multimode acceptance photon coincidence count setup. Subsequently, the results support the anticipated fact that down-converted radiation excited in $\chi^{(2)}$ media by continuous wave incoherent radiation is of non-classical origin and that this property is well preserved in the photon pair collection experiment.

4. Conclusions

In conclusion, photon coincidence experiment in SPDC radiation excited by an incoherent source was performed for the first time. We demonstrated that incoherently pumped spontaneous parametrical down-conversion delivers sufficient power for both single photons and coincidence counting using multimode acceptance photon counters. The guidable towards the detectors part of SPDC emission contained up to 49% biphoton states that cannot be

attributed to coincidences of the classical field. Results of a relatively simple semiclassical numerical model match well with the experiment, including coincidence probability calculations.

The results of the present research are auspicious for quantum experiments where nonlinear crystals pumped by incoherent light may be employed as a biphoton source. Its potential advantages are very low cost and design simplicity, as well as the high power scalability of a LED. Wavelength flexibility of the pump beam, thanks to variety of the commercially available LEDs, is also a factor not to be disregarded.

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